LA-UR-21-25357

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Title: EPC-ES WIPP WAC APPENDIX A EQUIVALENCE SUPPORT MEASUREMENTS 2019 - 2020

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Intended for: Annual (2019-2020) set of calibration confirmation measurements that serve to establish and demonstrate equivalency with the processes used by U.S. Department of Energy (DOE) Carlsbad Field Office (CBFO) certified nondestructive assay (NDA) systems.

Issued: 2021-06-07

EPC-ES WIPP WAC APPENDIX A
EQUIVALENCE SUPPORT MEASUREMENTS
2019 - 2020

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1.0 INTRODUCTION

Characterization of low-level waste (LLW) at Los Alamos National Laboratory (LANL) is primarily performed by the Environmental Protection and Compliance – Environmental Stewardship (EPC-ES) Characterization Services team and/or the Nuclear Process Infrastructure (NPI-9) Nuclear Material Support Services team (hereafter referred to as the gamma spectroscopy teams). The gamma spectroscopy teams use portable high-purity germanium (HPGe) detector systems for the nondestructive assay (NDA) characterization of LLW. Controlled-approved procedures and processes for the use of such systems to assay LLW items are currently in place. Additionally, a number of performance studies have been conducted by the gamma spectroscopy teams to support the efficacy and quality of assay results generated by the established NDA process.

LANL currently uses, and intends to continue to use, the Nevada National Security Site (NNSS) as an off-site disposal facility for some LLW. The NNSS issued a position paper (DOE/NV—1121, May 10, 2006) on certifying waste for NNSS disposal. This paper indicates that systems that are <u>not</u> certified by the U.S. Department of Energy (DOE) Carlsbad Field Office (CBFO) for Waste Isolation Pilot Plant (WIPP) disposal of transuranic (TRU) waste must demonstrate equivalent practices to the CBFO-certified systems in order for activity concentration values to be assigned to assayed items without adding in the total measurement uncertainty (TMU) i.e., using only the system's reported assay value. While most of the practices established for CBFO-certified systems are currently being executed within existing EPC-ES and NPI-9 processes and procedures, some supplementary NDA performance measurements are required to demonstrate full equivalence. This report defines these nominal measurements and presents the results of these measurements.

2.0 ANNUAL CALIBRATION CONFIRMATION AND PERFORMANCE CHECK MEASUREMENTS

Every portable NDA detector that the gamma spectroscopy teams use is calibrated for intrinsic efficiency and angular response when it is first procured and commissioned into service. Energy calibration is performed initially before the efficiency calibration and daily during periods of operation as part of the daily quality control (QC) checks specified in approved LANL operating procedures.

DOE/WIPP-02-3122, "Transuranic Waste Acceptance Criteria for the Waste Isolation Pilot Plant [WIPP WAC] Appendix A, Radioassay Requirements for Contact-Handled Transuranic Waste" (DOE 2013), specifies that verification of a NDA system calibration is performed if major system repairs or modifications occur. If a gamma spectroscopy team HPGe detector system undergoes a major system repair or modification, a new efficiency calibration is performed to replace the initial calibration. Thus, verification of the initial efficiency calibration is <u>not</u> applicable when system repairs or modifications occur. Also, as a best management practice, the gamma spectroscopy teams update the efficiency calibration for each of their detectors periodically. The angular response calibration for a particular detector/shield/collimator configuration is a mathematical function depicting the relative response the detector has for the range of angles with which the gamma rays could be entering the detector's field of view. This curve (for a particular detector/shield/collimator configuration) is dependent only on the size of the detector crystal and remains valid even if the efficiency of the detector is recalibrated.

WIPP WAC Appendix A specifies that the confirmation of each calibration or recalibration be performed by evaluating replicate measurements of a noninterfering matrix for accuracy and precision. Also, WIPP WAC Appendix A specifies that as part of performance checks, the long-term stability of the

NDA system matrix correction of an interfering matrix must be assessed. Surrogate waste containers used must reflect the type of waste (e.g., zero/empty or debris) currently being assayed.

With the modeling program SNAP™ (Spectral Nondestructive Assay Program), most of the elements normally associated with a WIPP-certified system calibration correction factor are mathematically calculated to produce a method-specific “calibration” each time: using the item-to-detector geometry, the matrix and filter attenuation losses, gamma-ray emission probability per decay, HPGe efficiency calibration, and field-of-view angular response. The only elements of this method-specific calibration that are unique to a particular HPGe detector and shield configuration are the HPGe intrinsic efficiency calibration and field-of-view angular response. All the other elements of the SNAP™ method-specific “calibration” are the same for all portable HPGe NDA detector systems.

Thus, a set of calibration confirmation measurements, using two of the gamma spectroscopy teams’ HPGe detector systems and representative of the full inventory of HPGe detectors, serves as a baseline to confirm the SNAP™ portable HPGe detector calibrations. The measurements were evaluated for accuracy (% recovery [%R]) and precision (% relative standard deviation [%RSD]) in accordance with WIPP WAC Appendix A and criteria set forth by CBFO, as applicable, in DOE/CBFO-01-1005, “Performance Demonstration Program Plan for Nondestructive Assay of Drummed Waste for the TRU Waste Characterization Program” (DOE 2015).

Further, in order to assess the long-term stability of the SNAP™ NDA process’s matrix correction of an interfering matrix, similar measurements – and accuracy and precision evaluations – will be done annually as performance check measurements. The annual performance check measurements will use a different subset of EPC-ES and NPI-9 HPGe detector systems each year until all HPGe detector systems in use have participated. Thereafter, the detector system selection cycle will repeat.

The two detectors used for the 2019 - 2020 measurements were serial numbers 4994529 and 4994514 (also referred to as OBIWAN and YODA, respectively). OBIWAN and YODA are Canberra BEGe HPGe detectors with a nominal 18% relative efficiency.

The measurements were targeted to satisfy two ranges of activity concentration loadings and two matrix types for each container type (55-gallon drum and ST-90 box). The target measurements are defined in Table 1, Annual Calibration Confirmation and Performance Check Measurements. This nominal set of measurements covers containers, matrices, and activity loadings being assayed in real LLW cases. The 55-gallon drum is the most common drum size used at LANL for waste packaging, and the ST-90 box is representative (by volume–nominal 90 cubic feet) of the majority of non-drum waste containers used at LANL. Radioactive source standards used to achieve the activity loadings were different from the radioactive source standards used for the intrinsic efficiency calibrations.

Several WIPP Performance Demonstration Program (PDP) plutonium source standards available at LANL (used for CBFO’s LANL TRU program) were used for the measurements. However, because the activity of the available sources was limited, the nominal total TRU alpha activity concentration targeted was not exactly achievable in most cases. The actual activity concentration loadings achieved are also listed in Table 1. The sources used for the measurements were of weapons-grade (WG) isotopic composition, with Pu-239 the dominant radionuclide present by mass.

Table 1. Annual Calibration Confirmation and Performance Check Measurements

Container	Matrix	Nominal Activity Concentration	Actual (WG Pu) Activity Concentration	Nominal Radioactive Material Distribution	Number of Replicates	*Nominal* Count Time
55-gallon Drum	Empty (Zero)	50 nCi/g	71 nCi/g	Uniform inside surface	6	900 s
		120 nCi/g	148 nCi/g			
	PDP Debris	50 nCi/g	65 nCi/g	Uniform distribution		
		120 nCi/g	134 nCi/g			
	PDP Sludge	80 nCi/g	96 nCi/g	Uniform distribution		
ST-90	Metal	50 nCi/g	53 nCi/g	Uniform distribution		3600 s
		120 nCi/g	119 nCi/g			
	Debris	50 nCi/g	58 nCi/g	Uniform distribution		
		120 nCi/g	122 nCi/g			

PDP like drums (55-gallon) similar to those used by the CBFO PDP program for official PDP testing were available for the measurements. The empty "Zero" matrix drum is identified as PDP matrix drum 001, and the debris matrix drum is identified as PDP matrix drum 003, with 33.1 kg and 36.5kg net weight, respectively. Aluminum source tubes 1, 2, and 3 were used to position PDP sources in the drums. The relative tube positions in the drum are identified in Figure 1, PDP drums radial tube placement and naming conventions. The PDP 003 debris drum used was built by the CBFO PDP program; the PDP 001 drum used was built by the EPC-ES team with the exact same tube make and configuration as the PDP 001 drum built by the CBFO PDP program. Detailed design specifications for the PDP drums can be found in DOE/CBFO-01-1005 (DOE 2015).

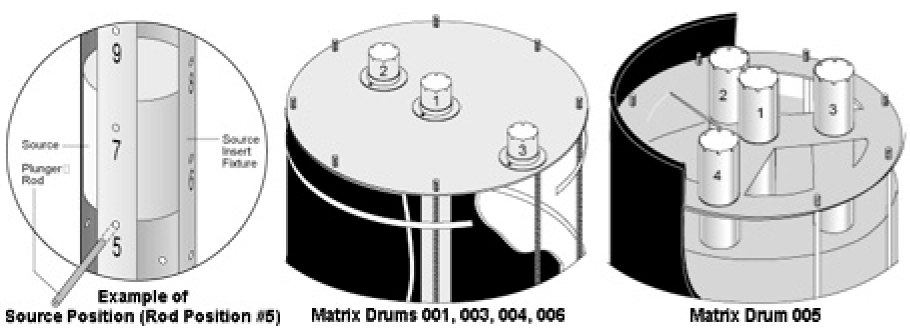

Figure 1. PDP drums radial tube placement and naming conventions

The quantitative radial positions of the tubes for PDP drums 001 and 003 are defined in Figure 2, Detailed radial positioning of the source tubes for PDP drums 001 and 003. Tube 1 is located at radius = 0 inches (the radial center of the drum) and is named R0; tube 2 is located at radius = 5.5 inches and is named R5.5; and tube 3 is located at radius = 9 inches and is named R9. All the sources were positioned with the center of the source at a height of 15.5 inches in the source tubes–the approximate vertical center height of the matrix material.

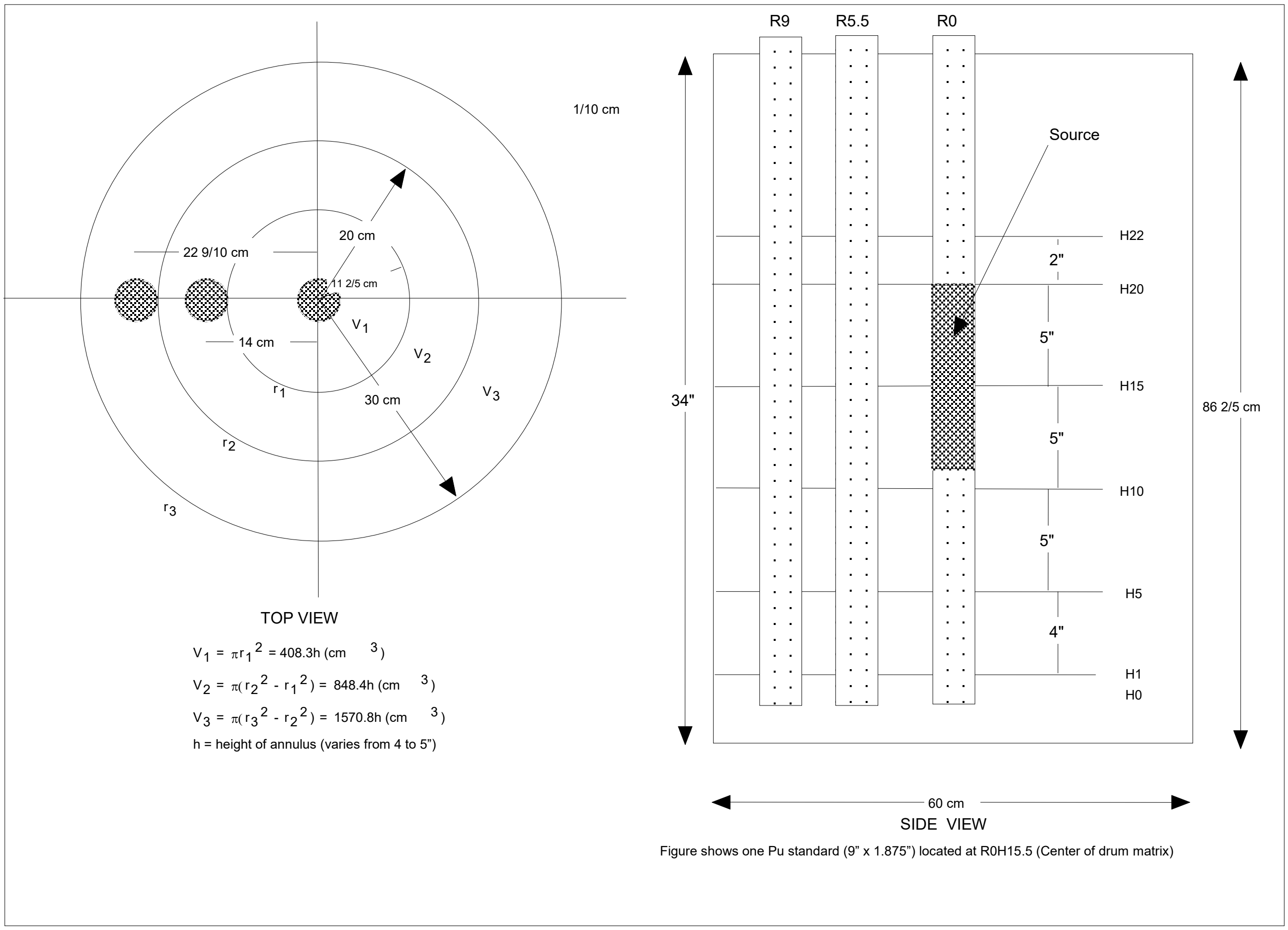

Figure 2. Detailed radial positioning of the source tubes for PDP drums 001 and 003

The PDP sludge matrix drum (identified as PDP matrix drum 005) built by the CBFO PDP program, with 177 kg of net matrix weight, was available for the sludge measurements. In the case of sludge waste, the radioactive material in the sludge mix is extremely likely to be uniformly distributed (especially along the radial coordinate). This expectation of uniform distribution is much stronger in the case of sludge than the debris matrix. The PDP drum 005 has four tubes as shown in Figure 1, PDP drum radial tube placement and naming conventions. Tube 1 is located at radius = 0 inches (the radial center of the drum) and is named R0; tube 2 is located at radius = 3.5 inches and is named R3.5; tube 3 is located at radius = 6 inches and is named R6; and tube 4 is located at radius = 9 inches and is named R9 The tube geometry, however, was not specifically designed to represent a uniform source distribution. Thus, the source placement for the annual calibration and performance check measurements were a best approximation of uniform distribution given the configured tube positions. The PDP matrix drum 005 was also used for the lower limit of detection (LLD) and TMU bounding measurements.

The %R and %RSD results for the PDP drum measurements were tabulated for Pu-239 mass and are shown in Table 2, Annual Calibration Confirmation and Performance Check Measurement Results for 55-gallon PDP Drums. The drum net weights and source tubes used to position the PDP sources are included in Table 2.

Table 2. Annual Calibration Confirmation and Performance Check Measurement Results for 55-Gallon PDP Drums

Container	Detector	Matrix (wt [kg])	WG Pu Activity Concentration	Source Tubes	Pu-239 Mass (g)	Average Pu-239 Measured Mass (g)	Average %R	%RSD
55-gallon Drum	OBIWAN	Zero (33.1)	71 nCi/g	R5.5	2.68E-02	3.15E-02	117.41	11.22
			148 nCi/g	R5.5	5.55E-02	6.52E-02	117.56	6.61
		Debris (36.5)	65 nCi/g	R5.5	2.68E-02	3.29E-02	122.89	13.96
			134 nCi/g	R5.5	5.55E-02	6.40E-02	115.31	6.60
		Sludge (177)	96 nCi/g	R6; R9	1.89E-01	2.18E-01*	115.41	16.70
	YODA	Zero (33.1)	71/g	R5.5	2.68E-02	2.99E-02	111.57	9.42
			148 nCi/g	R5.5	5.55E-02	5.97E-02	107.65	6.73
		Debris (36.5)	65 nCi/g	R5.5	2.68E-02	2.96E-02	110.32	12.85
			134 nCi/g	R5.5	5.55E-02	6.11E-02	110.08	11.63
		Sludge (177)	96 nCi/g	R6; R9	1.89E-01	1.98E-01*	104.81	14.51

* = 1800 seconds

For the ST-90 box measurements, the Characterization Services team constructed matrix boxes for two target matrix types – Debris and Metal – using clean scrap material from LANL. Two empty ST-90 boxes were obtained and seven (7) source tubes were installed in each box. The source tubes were aluminum with a 0.125-inch wall thickness. A top view of a ST-90 box illustrating the positions and naming of the source tubes is presented in Figure 3, Diagram of ST-90 box tube placement and naming convention. Tube T0 is located at the center of the box in each X and Y direction–T0 defines the X=0 and Y=0 position. Tubes T1 and T3 are 11.5 inches from T0 (center) directly along the Y axis. Tubes T2 and T4 are 11.5 inches from T0 (center) directly along the X axis. Tubes T5 and T6 are 18 inches from T0 (center) directly along the X axis.

Clean scrap material consisting primarily of mixed metal (steel, aluminum, and copper) pipes, tubes, and shelves were loaded into one box to make up the ST-90 metal matrix case. This type of material is typical in an actual metal waste box. A net weight of 944 kg was achieved for the ST-90 metal box. This weight was in the mid-range of a typical metal waste box weight.

Clean scrap cardboard, paper, and wood (generally referred to as combustible material) were loaded separately into the second box to make up the ST-90 debris matrix case. This type of combustible material is typical in an actual debris/combustible waste box. A net weight of 292 kg was achieved for the ST-90 debris box. This weight was in the low range of a typical debris/combustible waste box weight.

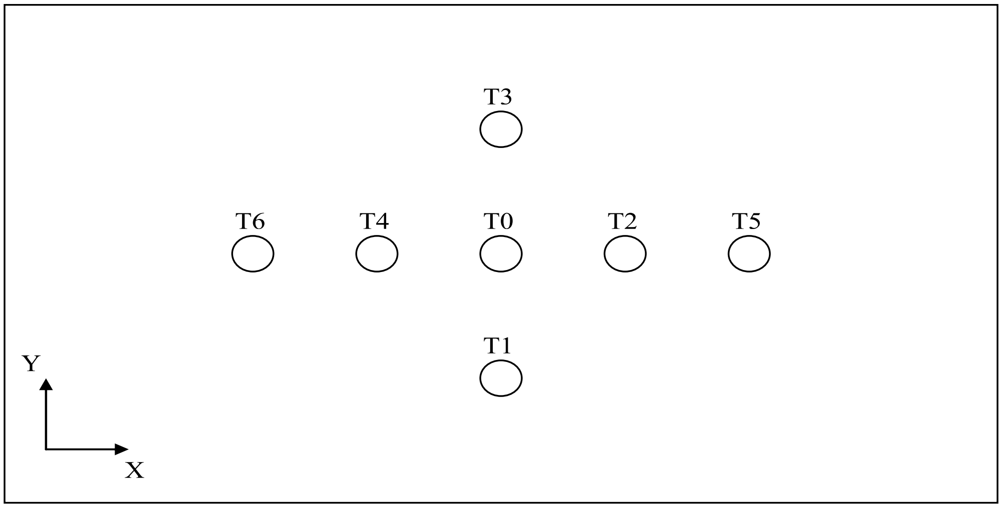

Figure 3. Diagram of ST-90 box tube placement and naming convention

The %R and %RSD results for the ST-90 box measurements were tabulated for Pu-239 mass and are shown in Table 3, Annual Calibration Confirmation and Performance Check Measurement Results for ST-90 Boxes. The box net weights and source tubes used to position the PDP sources are included in Table 3. All the sources were positioned with the center of the source at a height of 24 inches in the source tubes – the vertical center height.

Table 3. Annual Calibration Confirmation and Performance Check Measurement Results for ST-90 Boxes

Container	Detector	Matrix (wt [kg])	Activity Concentration	Source Tubes	Pu-239 Mass (g)	Average Pu-239 Measured Mass (g)	Average %R	%RSD
ST-90	OBIWAN	Metal (944)	53 nCi/g	T2 & T4	5.74E-01	6.22E-01	108.35	10.15
			119 nCi/g	T0, T2, & T4	1.23E+00	1.18E+00	95.62	6.27
		Debris (292)	58 nCi/g	T1 & T3	1.89E-01	2.15E-01	113.73	13.86
			122 nCi/g	T0, T2, & T4	3.93E-01	3.93E-01	100.15	4.69
	YODA	Metal (944)	53 nCi/g	T2 & T4	5.74E-01	6.07E-01	105.74	6.60
			119 nCi/g	T0, T2, & T4	1.23E+00	1.23E+00	100.09	7.44
		Debris (292)	58 nCi/g	T1 & T3	1.89E-01	2.08E-01	110.02	13.29
			122 nCi/g	T0, T2, & T4	3.93E-01	3.94E-01	100.36	8.75

3.0 LLD MEASUREMENTS

WIPP WAC Appendix A specifies that the lower limit of detection (LLD) for each radioassay system must be determined. Instruments performing TRU/LLW discrimination measurements must have a LLD of 100 nCi/g or less (total TRU alpha activity concentration). This is more correctly referred to as the minimum detectable concentration (MDC). The LLD for SNAP™-based HPGe systems has been determined and documented by LANL for Pu-239 (Myers et al, 2001). To demonstrate the LLDs for all the WIPP-tracked nuclides that contribute to the total TRU alpha activity, replicate measurements were made on the range of containers and matrices being assayed in real LLW cases. The target measurements that were performed are defined in Table 4, LLD Measurements (One Time). The LLD measurements were performed for the two HPGe detector systems OBIWAN and YODA for the same PDP drums and ST-90 boxes used for the calibration confirmation and performance check measurements.

Table 4. LLD Measurements (One Time)

<table>
<tr><th>Container</th><th>Matrix</th><th>Activity Loadings</th><th>Number of Replicates</th><th>Nominal Count Time</th></tr>
<tr><td rowspan="3">55-gallon Drum</td><td>Empty</td><td rowspan="5">0</td><td rowspan="5">6</td><td rowspan="3">900 s</td></tr>
<tr><td>PDP Debris</td></tr>
<tr><td>PDP Sludge</td></tr>
<tr><td rowspan="2">ST-90</td><td>Debris</td><td rowspan="2">3600 s</td></tr>
<tr><td>Metal</td></tr>
</table>

LLD values in units of nCi/g (MDCs) were tabulated for all of the 10 WIPP-tracked radionuclides and U-235. The MDCs are based on the gamma ray for each radionuclide providing the optimum MDC (except for Am-241, where the 59.5-keV gamma is optimum but not generally used because of its low energy). The MDC results for the detector OBIWAN for the PDP drums are shown in Table 5, while the MDC results for YODA for the PDP drums are presented in Table 6.

Table 5. MDC Results for OBIWAN for the PDP Drums

OBIWAN–PDP Drums												
Matrix	**Matrix Mass (kg)**	**MDC |(keV)**	**Am-241** 125.29	**Cs-137** 661.65	**Pu-238** 99.86	**Pu-239** 129.29	**Pu-240** 160.28	**Pu-242** 158.80	**U-233** 317.13	**U-234** 120.91	**U-235** 185.72	**U-238** 1001.00
Zero	33.1	Average (nCi/g)	1.55E+01	1.12E-03	1.03E+01	1.01E+01	1.39E+02	1.25E+02	7.78E+00	1.93E+00	1.05E-03	1.43E-01
		STD (nCi/g)	3.49E-01	1.11E-04	3.66E-01	3.27E-01	4.46E+00	1.79E+00	4.44E-01	6.50E-02	4.56E-05	2.10E-02
		%RSD	2.25%	9.89%	3.54%	3.23%	3.20%	1.43%	5.71%	3.37%	4.34%	14.69%
Debris	36.5	Average (nCi/g)	2.48E+01	1.29E-03	1.80E+01	1.61E+01	2.11E+02	1.99E+02	1.09E+01	3.22E+00	1.60E-03	1.68E-01
		STD (nCi/g)	1.05E+00	1.33E-04	5.82E-01	5.64E-01	5.09E+00	8.52E+00	6.35E-01	7.33E-02	4.86E-05	1.40E-02
		%RSD	4.23%	10.31%	3.24%	3.50%	2.41%	4.29%	5.80%	2.28%	3.04%	8.36%
Sludge	177	Average (nCi/g)	1.71E+01	6.58E-04	1.21E+01	1.10E+01	1.40E+02	1.26E+02	6.18E+00	2.13E+00	9.77E-04	7.69E-02
		STD (nCi/g)	3.61E-01	3.02E-05	4.36E-01	3.31E-01	7.96E+00	2.97E+00	6.60E-01	4.67E-02	2.98E-05	4.96E-03
		%RSD	2.11%	4.59%	3.60%	3.01%	5.69%	2.35%	10.68%	2.19%	3.05%	6.45%

Table 6. MDC Results for YODA for the PDP Drums

YODA–PDP Drums												
Matrix	**Matrix Mass (kg)**	**MDC |(keV)**	**Am-241** 125.29	**Cs-137** 661.65	**Pu-238** 99.86	**Pu-239** 129.29	**Pu-240** 160.28	**Pu-242** 158.80	**U-233** 317.13	**U-234** 120.91	**U-235** 185.72	**U-238** 1001.00
Zero	33.1	Average (nCi/g)	1.45E+01	9.98E-04	1.00E+01	9.47E+00	1.26E+02	1.17E+02	7.15E+00	1.80E+00	9.86E-04	1.25E-01
		STD (nCi/g)	3.98E-01	1.03E-04	3.87E-01	3.08E-01	5.98E+00	4.15E+00	4.93E-01	3.39E-02	2.97E-05	8.94E-03
		%RSD	2.75%	10.29%	3.86%	3.26%	4.74%	3.54%	6.90%	1.88%	3.01%	7.13%
Debris	36.5	Average (nCi/g)	2.45E+01	1.29E-03	1.71E+01	1.54E+01	2.03E+02	1.84E+02	1.10E+01	3.01E+00	1.49E-03	1.61E-01
		STD (nCi/g)	5.76E-01	1.33E-04	3.58E-01	5.85E-01	5.04E+00	5.67E+00	5.59E-01	8.89E-02	6.28E-05	1.32E-02
		%RSD	2.35%	10.32%	2.09%	3.79%	2.48%	3.09%	5.08%	2.95%	4.21%	8.18%
Sludge	177	Average (nCi/g)	1.62E+01	5.93E-04	1.18E+01	1.04E+01	1.31E+02	1.17E+02	5.50E+00	2.07E+00	9.02E-04	6.87E-02
		STD (nCi/g)	8.27E-01	3.62E-05	3.19E-01	1.55E-01	3.06E+00	2.88E+00	4.56E-01	2.66E-02	2.98E-05	8.53E-03
		%RSD	5.11%	6.10%	2.71%	1.49%	2.34%	2.46%	8.30%	1.29%	3.30%	12.42%

The ST-90 Box MDC results for the detectors OBIWAN and YODA are shown in Table 7 and Table 8, respectively.

Table 7. MDC Results for OBIWAN for the ST-90 Boxes

OBIWAN–ST-90 Box												
			Am-241	**Cs-137**	**Pu-238**	**Pu-239**	**Pu-240**	**Pu-242**	**U-233**	**U-234**	**U-235**	**U-238**
Matrix	**Matrix Mass (kg)**	**MDC |(keV)**	125.29	661.65	99.86	129.29or 413.70	160.28	158.80	317.13	120.91	185.72	1001.00
Debris	292	Average (nCi/g)	6.32E+00	2.67E-04	4.95E+00	4.05E+00	5.09E+01	4.58E+01	2.43E+00	8.01E-01	3.65E-04	3.59E-02
		STD (nCi/g)	1.73E-01	7.22E-06	7.18E-02	5.29E-02	1.06E+00	1.02E+00	3.92E-02	1.02E-02	8.10E-06	1.62E-03
		%RSD	2.73%	2.71%	1.45%	1.31%	2.07%	2.23%	1.61%	1.28%	2.22%	4.51%
Metal	944	Average (nCi/g)	7.34E+00	1.43E-04	7.82E+00	4.56E+00	4.22E+01	3.90E+01	1.47E+00	9.77E-01	2.75E-04	1.64E-02
		STD (nCi/g)	1.12E-01	7.87E-06	9.82E-02	1.15E-01	9.87E-01	8.75E-01	4.45E-02	1.65E-02	5.99E-06	8.22E-04
		%RSD	1.52%	5.51%	1.26%	2.52%	2.34%	2.24%	3.02%	1.69%	2.18%	5.01%

Table 8. MDC Results for YODA for the ST-90 Boxes

YODA–ST-90 Box												
			Am-241	**Cs-137**	**Pu-238**	**Pu-239**	**Pu-240**	**Pu-242**	**U-233**	**U-234**	**U-235**	**U-238**
Matrix	**Matrix Mass (kg)**	**MDC |(keV)**	125.29	661.65	99.86	129.29 or 413.70	160.28	158.80	317.13	120.91	185.72	1001.00
Debris	292	Average (nCi/g)	6.10E+00	2.32E-04	4.81E+00	3.92E+00	4.78E+01	4.27E+01	2.32E+00	7.75E-01	3.45E-04	2.95E-02
		STD (nCi/g)	7.69E-02	1.13E-05	8.41E-02	3.58E-02	9.22E-01	6.77E-01	7.23E-02	1.17E-02	3.87E-06	1.96E-03
		%RSD	1.26%	4.88%	1.75%	0.91%	1.93%	1.59%	3.12%	1.51%	1.12%	6.64%
Metal	944	Average (nCi/g)	6.90E+00	1.34E-04	7.52E+00	4.23E+00	3.98E+01	3.70E+01	1.42E+00	9.34E-01	2.58E-04	1.43E-02
		STD (nCi/g)	9.99E-02	4.43E-06	1.70E-01	5.61E-02	6.66E-01	4.00E-01	7.63E-02	2.10E-02	4.59E-06	9.99E-04
		%RSD	1.45%	3.30%	2.26%	1.33%	1.67%	1.08%	5.36%	2.24%	1.78%	6.97%

4.0 TMU BOUNDING MEASUREMENTS

In SNAP™, the combined geometry and attenuation (G&A) component of the TMU is calculated for the 95th percentile worst location in the assayed container. Consequently, for some assayed waste cases, SNAP™ calculates large TMU estimates. In most of these cases, the radioactive material is <u>not</u> likely to be located in the 95th percentile worst position on the container. Thus, for such cases, the 95th percentile G&A error is an overestimate and is <u>not</u> used for final reporting. Instead, the expert analyst will reduce the error based on historic studies and experience of what error estimate would better represent a real-world waste item with several dispersed sources of radioactive material in heterogeneous matrices or uniform activity spread throughout the waste matrix in homogenous matrices. Several LANL technical papers provide a basis for error reduction in many such cases. However, in order to better establish specific guidelines for the expert SNAP™ analyst to achieve the best uncertainty reporting, additional measurements were performed to further determine the effects of source positioning on assay results and establish TMU bounds for error reduction. Replicate measurements were performed on the same range of containers and matrices evaluated for calibration confirmation and performance checks. The nominal target measurements are defined in Table 9, TMU Bounding Measurements (One Time). Again, because the activity of the available sources was limited, the nominal total TRU alpha activity concentration targeted was not exactly achievable in most cases. The actual WG plutonium activity concentration loadings achieved are also listed in Table 9. Again, most of the sources used for the measurements were of WG isotopic composition.

Table 9. TMU Bounding Measurements (One Time)

<table>
<tr><th>Container</th><th>Matrix</th><th>Nominal Activity Concentration</th><th>Actual Activity Concentration</th><th>Nominal Radioactive Material Distribution</th><th>Number of Replicates</th><th>Nominal Count Time</th></tr>
<tr><td rowspan="6">55-gallon Drum</td><td rowspan="2">Empty</td><td rowspan="2">80 nCi/g</td><td rowspan="2">77 nCi/g</td><td>Center/Center</td><td rowspan="10">6</td><td rowspan="6">900 s</td></tr>
<tr><td>Uniform Distribution</td></tr>
<tr><td rowspan="2">Debris</td><td rowspan="2">80 nCi/g</td><td rowspan="2">70 nCi/g</td><td>Center/Center</td></tr>
<tr><td>Uniform Distribution</td></tr>
<tr><td rowspan="2">Sludge</td><td rowspan="2">120 nCi/g</td><td rowspan="2">126 nCi/g</td><td>Center/Center</td></tr>
<tr><td>Outer Ring</td></tr>
<tr><td rowspan="4">ST-90</td><td rowspan="2">Debris</td><td rowspan="2">80 nCi/g</td><td rowspan="2">75 nCi/g</td><td>Center/Center</td><td rowspan="4">3600 s</td></tr>
<tr><td>Uniform Distribution</td></tr>
<tr><td rowspan="2">Metal</td><td rowspan="2">80 nCi/g</td><td rowspan="2">79 nCi/g</td><td>Center/Center</td></tr>
<tr><td>Uniform Distribution</td></tr>
</table>

PDP sources and drums (debris and zero matrices) were also used for the TMU bounding measurements. The %R and %RSD results for the PDP drum measurements were tabulated for Pu-239 mass and are shown in Table 10, TMU Measurement Results for 55-gallon PDP Drums. The drum net weights and source tubes used to position the PDP sources are included in Table 10. All the sources were positioned with the center of the source at a height of 15.5 inches in the source tubes – the vertical center height.

Table 10. TMU Measurement Results for 55-gallon PDP Drums

Container	Detector	Matrix and (wt [kg])	Activity Concentration	Source Tubes	Pu-239 Mass (g)	Average Pu-239 Measured Mass (g)	Average %R	%RSD
55-gallon Drum	OBIWAN	Zero (33.1)	77 nCi/g	R5.5	2.88E-02	3.54E-02	122.93	8.31
				R0	2.88E-02	3.64-02	126.52	15.06
		Debris (36.5)	70 nCi/g	R5.5	2.88E-02	3.21E-02	111.57	13.86
				R0	2.88E-02	2.70E-02	93.91	18.09
		Sludge (177)	126 nCi/g	R9	2.43E-01	3.61E-01	148.24	14.03
				R0	2.43E-01	4.38E-02	18.01	16.79
	YODA	Zero (33.1)	77 nCi/g	R5.5	2.88E-02	3.50E-02	121.60	9.12
				R0	2.88E-02	3.52E-02	122.23	13.80
		Debris (36.5)	70 nCi/g	R5.5	2.88E-02	3.37E-02	117.19	11.06
				R0	2.88E-02	2.71E-02	94.31	14.63
		Sludge (177)	126 nCi/g	R9	2.43E-01	3.88E-01	159.34	15.26
				R0	2.43E-01	5.50E-02	22.60	14.47

The constructed metal matrix and debris matrix ST-90 boxes were also used for the TMU bounding measurements. The %R and %RSD results for the ST-90 box measurements were tabulated for Pu-239 mass and are shown in Table 11, TMU Measurement Results for ST-90 Boxes. The box net weights and source tubes used to position the PDP sources are included in Table 11. All the sources were positioned with the center of the source at a height of 24 inches in the source tubes – the vertical center height.

Table 11. TMU Measurement Results for ST-90 Boxes

Container	Detector	Matrix (wt [kg])	Activity Concentration	Source Tubes	Pu-239 Mass (g)	Average Pu-239 Measured Mass (g)	Average %R	%RSD
ST-90	OBIWAN	Debris (292)	75 nCi/g	T1, T2, T3 & T4	2.44E-01	2.35E-01	96.39	12.01
				T0 & T1	2.44E-01	2.46E-01	100.89	10.84
		Metal (944)	79 nCi/g	T1 & T3	8.25E-01	1.42E+00	172.26	8.16
				T0	8.25E-01	1.02E+00	123.77	10.37
	YODA	Debris (292)	75 nCi/g	T1, T2, T3 & T4	2.44E-01	2.09E-01	85.60	8.83
				T0 & T1	2.44E-01	2.33E-01	95.36	11.49
		Metal	79 nCi/g	T1	8.25E-01	1.39E+0	168.83	10.88

		(944)		&T3				
				T0	8.25E-01	1.05E+00	127.31	9.99

5.0 DISCUSSION

5.1 Annual Calibration Confirmation and Performance Check Measurements

The annual calibration confirmation and performance check measurements were evaluated against the scoring criteria set forth by the WIPP PDP program (DOE 2015). The scoring criteria for the PDP program are divided into two waste matrix categories: Non-interfering and Interfering. The Non-interfering matrix category is applicable to the Zero matrix, and the Interfering matrix category is applicable to the Debris and Metal matrices. Further, the PDP scoring criteria are divided into activity range categories: Low, Mid-Low, Mid-High, and High. All of the drum measurements were in the Low activity range category. All of the box measurements were in the Mid-Low activity category, except for the Debris 58 nCi/g loading, which was in the Low activity range category. The detailed PDP scoring criteria are presented in Table 12, NDA PDP Activity Ranges and Associated Scoring Acceptance Criteria.

For all 55-gallon drum Zero matrix cases, the average %R was well within the allowed range of 70% to 130% (actual results from 107.65% to 117.56%), and the average %RSD was within the allowed maximum of 14% (actual results from 6.61% to 11.22%). For all 55-gallon drum Debris matrix cases, the average %R was well within the allowed range of 40% to 160% (actual results from 110.08% to 122.89%), and the average %RSD was within the allowed maximum of 16% (actual results from 6.60% to 13.96%). For the 55-gallon drum Sludge matrix case 1800 second counts were performed to improve counting statistics. Here the average %R was well within the allowed range of 40% to 160% (actual results from 104.81% to 115.41%), and the average %RSD for Yoda was within the allowed maximum of 16% (actual results from 14.51%). For Obiwan the %RSD was slightly over the 16% target coming in at 16.70%. Both Obiwan and Yoda are low efficiency detectors (nominal 15%) and it is believed that the slightly high %RSD is due to non-ideal counting statistics even with 1800 second counts.

For all ST-90 box Debris matrix cases, the average %R was well within the allowed range of 40% to 160% (actual results from 100.15% to 113.73%). For the Low range Debris matrix case, the average %RSD was within the allowed maximum of 16% (actual results from 13.29% to 13.86%). For the Mid-Low range Debris matrix case, the average %RSD was within the allowed maximum of 12% (actual results from 4.69% to 8.75%). For all ST-90 box Metal matrix cases, the average %R was within the allowed range of 40% to 160% (actual results from 95.62% to 108.35%), and the average %RSD was within the allowed Mid-Low maximum of 12% (actual results from 6.27% to 10.15%). The 413-keV Pu-239 gamma ray was used in the analysis of the ST-90 box data because of the increased volume (and density in the case of the metal) limiting the penetration of the 129-keV gamma rays through the box.

Table 12. NDA PDP Activity Ranges and Associated Scoring Acceptance Criteria

		Maximum Measured Precision[a]		Bias Range (%RL[b] and % RU[c])[d]			
Activity Range	**Range of Sample Activity (in α-curies[e])**	**Non-interfering Matrix (%RSD)**	**Interfering Matrix (%RSD)**	**Non-interfering Matrix (%R)**		**Interfering Matrix (%R)**	
Low	> 0 to 0.02	14	16	Lower:	70	Lower:	40
				Upper:	130	Upper:	160
Mid-Low	> 0.02 to 0.2	10.5	12	Lower:	70	Lower:	40
				Upper:	130	Upper:	160
Mid-High	> 0.2 to 2.0	7	12	Lower:	70	Lower:	40
				Upper:	130	Upper:	160
High	> 2.0	3.5	6	Lower:	70	Lower:	40
				Upper:	130	Upper:	160

[a] Measured precision that must be met to satisfy the precision criteria at the 95% upper confidence bound, based on six replicates. The values are one relative standard deviation referenced to the known value for the test.

[b] %RL = percent recovery, lower.

[c] %RU = percent recovery, upper.

d %RL and %RU values used in Equation 3 (DOE 2015) to determine the 95% confidence bound for the ratio of the mean of the measured values to the known value, expressed as a percent.

e Applicable range of TRU activity contained in a PDP sample; units are curies of alpha-emitting TRU isotopes with half-lives greater than 20 years.

5.2 LLD Measurements

The goal of the LLD measurements is to establish that the detectors used by the gamma spectroscopy teams have a LLD (or MDC) of 100 nCi/g or less (total TRU alpha activity concentration). The LLD is that level of radioactivity which, if present, yields a measured value greater than the critical level with a 95% probability, where the critical level is defined as that value which measurements of the background will exceed with 5% probability. The MDC calculation algorithm in SNAP™ is consistent with this definition. For completeness, MDC values for all 10 WIPP-tracked radionuclides plus U-235 were calculated and tabulated for each of the waste matrix/container combinations. However, the primary nuclide of interest here is Pu-239. This is because acceptable knowledge about the isotopic composition of the plutonium is generally used during characterization of plutonium waste at LANL. A common isotopic composition at LANL is WG plutonium, where Pu-239 accounts for 94% of the total plutonium mass – applicable to the PDP sources. The Pu-239 activity is generally directly measured, and the activity of the other plutonium isotopes and Am-241 is determined by correlation to the Pu-239 activity. The plutonium isotopes and americium account for all of the reportable TRU alpha activity in WG plutonium, so in this case the MDC for Pu-239 drives the MDC for WIPP-equivalent characterization.

For all cases, the MDC for Pu-239 for the 55-gallon drum measurements was < 17 nCi/g. For the Zero matrix, the Pu-239 MDC was $\leq$ 11 nCi/g, which is consistent with MDCs typically calculated for actual empty waste drums at LANL – i.e., drums that had waste in them but were subsequently emptied and have potential contamination. The MDC for Pu-239 for the ST-90 box measurements was < 5 nCi/g for the Debris matrix and < 5 nCi/g for the Metal matrix using the 129-keV peak. These improved MDCs for Debris compared with the 55-gallon drum MDCs reflect the increased matrix weight of the ST-90 boxes.

5.3 TMU Bounding Measurements

The main goal for the TMU bounding measurements was to compare nominal "bad source location" measurement data against nominal uniform distribution measurements data using the same activity loading, in order to aid in reducing reported uncertainties (from the SNAP™ default uncertainty estimates) when necessary.

For the 55-gallon Zero matrix drum case, the two %R results (uniform and center) were <1% to 4% different for both detectors, OBIWAN (127% and 123%) and YODA (122% and 122%). This is not all that unexpected, as the center tube source geometry (R0) is not appreciably much different from the R5 tube geometry when the drum is being rotated and only geometry (vs matrix) is the only differentiating factor. For the Debris matrix drum case, the %R for OBIWAN was 18% lower for the R0 source location (112% and 94%) and for YODA was 23% lower (117% and 94%). This is a trend one would normally expect (larger deviation) when a combustible matrix is present because matrix attenuation and geometry differences are both differentiating factors. Note that in the Debris matrix counts for the center location at 900 seconds show slightly poorer counting statistics (compared to the uniform location), with the %RSD ranging from 14.63% to 18.09%. This indicates that 900 seconds is a marginal count time for good counting statistics. In field practice an 1800 second minimum count time for a 55-gallon drum is more typical and would yield better counting statistics – 900 seconds was used as a nominal count time for the test TMU cases to establish a worst case.

For the 55-gallon Sludge matrix drum case, the two most extreme non-uniform cases were tested, R0 and R9, because a uniform distribution loading was not achievable with the Pu standards available. These two cases serve to bound the potential errors for a sludge drum measurement. As expected, the uniform distribution model results for the R0 loading showed a significant low bias with %R = 18% for OBIWAN and %R = 23 for YODA. The R9 loading as expected showed a high bias with %R = 148% for OBIWAN and %R = 159% for YODA.

For the ST-90 box Debris matrix case, expected small deviation between the uniform and center locations was observed. The %R for the T0 source positioning data for OBIWAN was 4% more and for YODA was 9% more than the T1/T2/T3/T4 source positioning data, respectively. All of these results for the Debris matrix case are well within the PDP program's allowed 40% to 160% %R range; furthermore, the %RSD values were also within the PDP program's allowed 12% range.

For the ST-90 box Metal matrix case, both T1/T3 location %R results were outside the PDP program's allowed 40% to 160% %R range (Obiwan 169% and Yoda 172%); the T0 location %R results were within the limits (Obiwan 124% and Yoda 127%) . All the T0 AND T1/T3 location %RSD values were within the PDP program's allowed range of 12%. The %R values revealed an unexpected pattern for the ST-90 box metal cases that is believed to be attributable to the density of metal, and a non-homogenous distribution of the metals including void spaces in the Metals ST-90. The T0 source positioning was expected to be a low-bias bad-case scenario. However, the %R for the T0 source positioning was 124% for OBIWAN and 127% for YODA, both showing no low bias of the source activities. Whereas the %R for the T1/T3 source positioning was 169% for OBIWAN and 172% for YODA, both showing a clear high bias of the source activity, both about 10% above the 160% upper PDP range. This bias is believed to be due to a combination of the positioning of the PDP sources in the box, the high density of metal (approximately 7.2 g/cm^3), and the increased volume of the ST-90 box. Two sources, of different amounts, were positioned in tubes T1 and T3. The model used to process the assay data assumed a uniform distribution of radioactive material in the box. However, for the Metal matrix, where a large correction is being applied because of the high nominal density of metal and the large volume of the ST-90 box, the model overcorrected for this source positioning loading – effectively, the gamma rays weren't encountering as much metal as the model predicted. This suggests that in-homogeneities in the Metals matrix including void spaces of air make the center "point source" source loading more representative of actual uniform distribution than the T1/T3 loading.

6.0 CONCLUSION

In conclusion, the EPC-ES and NPI-9 gamma spectroscopy teams at LANL have defined and performed measurements that serve to establish and demonstrate equivalency with the processes used by CBFO-certified NDA systems. The supplemental measurements address four key areas in Appendix A of the WIPP WAC: Annual calibration confirmation, performance check measurements; LLD determination; and TMU definition. For these measurements the containers, matrices and activity loadings are selected to represent items being assayed in real LLW cases. The LLD and the TMU bounding measurements are to be performed one time per detector and will <u>not</u> be required to be repeated in future campaigns. The NNSS annual calibration and performance check measurements were performed initially and are planned to repeat in annual campaigns in order to maintain NNSS certification. PDP sources and drums were used for the measurements, and ST-90 matrix boxes were constructed specifically for this exercise. In all the calibration and performance check cases, the results for accuracy and precision (%R and %RSD, respectively) were well within allowable ranges as defined by the WIPP PDP program (DOE 2015). LLD (or MDC) results were established for all of the 10 WIPP-reportable radionuclides and U-235, and the MDC for Pu-239 was established in all cases to be well under 100 nCi/g. Finally, useful results for reducing estimated total measurement uncertainties were established, and interesting expected and unexpected cases of bias differences between source loadings were observed and will be applied toward this end.

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